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(54) **Herbicide microemulsion concentrate**

(57) A stable water-dispersible herbicide microemulsion concentrate composition contains as the active ingredient a water-insoluble herbicide, such as fluroxypyr-1-methylheptyl ester, combined with a water-soluble salt of N-phosphonomethyl-glycine, for example the isopropylamine salt; together with at least one amine ethoxylate containing 2 to 5 moles of ethylene oxid ; at least one amine ethoxylate surfactant containing from 8 to 20 moles of ethylene oxide; and at least one cosurfactant which is a glycol ether, a C₄-C₁₂ aliphatic alcohol, an alcohol ether based on ethylene, propylene or butylene oxide, or a nonionic surfactant with an HLB value of less than 10.

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HERBICIDAL AQUEOUS-BASED MICROEMULSION COMPOSITIONS

The present invention relates to novel oil-in-water microemulsions of herbicidal chemicals and methods
5 for preparing and using said microemulsions for controlling undesirable plant species.

It is a practice within agriculture directed to the growing of crop plants to use a range of products
10 containing a variety of active ingredients to obtain a broad spectrum of control of undesirable plant species. When applying two or more products to a crop, preparation containing two or more active ingredients are usually mixed in a tank shortly before their field application.
15 Such mixtures are often referred to as tank-mixtures and normally contain two or more active ingredients where the combined spectra of activity makes up for any deficiencies of each of the individual components.

20 Tank-mixtures suffer from the disadvantage in that the farmer must purchase and store each separate herbicide until the actual preparation of such tank-mixture in the field. The farmer is also required to measure out varying amounts of each different
25 herbicide used in the tank-mixture allowing for the possibility of mixing errors. Accordingly, a pre-packaged mixture, i.e., concentrate, containing a mixture of herbicides in one container is highly desirable since the farmer only has to purchase and store the single pre-
30 packaged mixture containing the active ingredients and the possibility of mixing errors is eliminated. An appropriate amount of the prepackaged concentrate can be added directly to the tank used for application, and the concentrate diluted with the addition of water for the in
35 situ preparation of the tank-mixture.

However, difficulties in preparing a prepackaged concentrate occurs when one of the herbicides is immiscible with water, or when the composition contains a mixture of herbicides, which are not both miscible in the same desired solvent. To overcome solvent incompatibility, a concentrated emulsion formulation can be used. A concentrated emulsion formulation consists of two phases and is a dispersion of one immiscible liquid (the discontinuous phase) in a second liquid (the continuous phase). If the continuous phase is water, the emulsion is an oil-in-water type emulsion and the water-immisible liquid or solution is generally referred to as the "oil" regardless of its nature. In a concentrated emulsion formulation the active ingredient is usually dissolved in an organic solvent along with added emulsifiers or dispersants. When the concentrate is added to water, the active ingredient becomes dispersed throughout the water.

Formulations have also been developed where a sparingly water-soluble herbicide or mixture of such herbicides is dissolved within the oil phase and a water-soluble herbicide is carried within the aqueous phase. A water-dispersible herbicide emulsion composition containing the water-soluble herbicide N-phosphonomethylglycine in the aqueous phase and the water-insoluble herbicide 2-haloacetanilide herbicide in the oil phase is disclosed in U.S. Patent 4,440,562. The emulsifier system in such a formulation is an anionic-nonionic blend containing alkylaryl sulfonate, phosphate esters of nonylphenol ethoxylates and polyalkyleneglycol ethers.

The formation of an emulsion, where the particle size is generally greater than 0.4 microns and

opaque to light, requires the input of a considerable amount of mechanical energy in the form of high shear mixing. Such emulsions are kinetically stable due to the mechanical energy required to form the emulsion. To
5 prevent the emulsion from separating into separate phases, gels or thickening surfactants are added.

Microemulsions, on the other hand, present a unique class of thermodynamically-stable liquid
10 dispersions where the dispersed particles are less than 0.1 microns in diameter. Microemulsions are transparent to light and have a low interfacial tension which makes microemulsions thermodynamically stable without the need to add a large amount of mechanical energy to form the
15 microemulsion, or addition of gels to thicken and stabilize it. To achieve the low interfacial tension necessary to form a microemulsion, they are generally prepared with more than one surfactant or with a mixture of a surfactant and a cosurfactant (e.g., a polar
20 compound of intermediate chain length). This combination of surfactants is usually required to provide the proper balance between hydrophilic and lipophilic properties (HLB, hydrophile- lipophile balance) for the required oil and water phases.

25 Although the formation of microemulsions is obtainable using certain surfactant combinations and finite concentrations of these combinations, formulating herbicide microemulsions is still an art. For example,
30 the choice of solvents and emulsifiers should be chosen to keep the herbicides in solution in the concentrate even at low temperatures and emulsifiable when diluted in water. In the preparation of a microemulsion formulation it is also desirable that the herbicide which is soluble
35 in the aqueous phase should be essentially insoluble in

the organic phase. Conversely, the herbicide that is soluble or suspended in the organic solvent should be essentially insoluble in the aqueous phase. This is necessary to ensure that over the period of shelf-life of a concentrate product, that the concentrate remain physically stable. Should the water solubility be excessive, crystal growth will occur in the concentrate leading to physical disruption of the concentrate.

Additional difficulties are encountered in the preparation of microemulsions when it is desirable to place active ingredients within both the oil and water phases. When preparing such a combination concentrate product, it is necessary to develop a system containing an appropriate surfactant, or blend of surfactants, and a cosurfactant to maintain the stability of the microemulsion and to maintain the stability of the active ingredients within the two phases.

The present invention is directed to a water-dispersible herbicidal microemulsion composition comprising:

A. from 1 to 30 percent by weight of at least one water insoluble herbicide;

B. optionally, from 0 to 25 percent by weight of a non water-miscible organic solvent in which the water insoluble herbicide is soluble;

C. from 1 to 30 percent by weight of a water-soluble salt of glyphosate;

D. from 0.5 to 35, preferably from 0.5 to 10 percent by weight of at least one amine ethoxylate surfactant containing from 2 to 5 moles of ethylene oxide;

E. from 0.5 to 35, preferably from 0.5 to 10 percent by weight of at least one amine ethoxylate surfactant containing from 8

to 20 moles of ethylene oxide;

F. from 0.5 to 10 percent by weight of at least one cosurfactant which is a glycol ether, a C₄-C₁₂ aliphatic alcohol, an alcohol ether based on ethylene, 5 propylene or butylene oxide, or a nonionic surfactant with an HLB value of less than 10; and,

G. sufficient water to serve as the continuous phase for the emulsion.

10 The term "herbicide" is used herein to mean an active ingredient which controls or adversely modifies the growth of plants because of phytotoxic or other effects substantial enough to seriously retard the growth of the plant or further to damage the plant sufficiently 15 to kill the plant.

The term "stability" refers to the resistance of emulsions to the coalescence of their dispersed droplets.

20

The term "plants" is meant to include germinant seeds, emerging seedlings, rhizomes and established vegetation.

25 The term "water-insoluble" as used herein means having a solubility in water of less than about 0.1 percent by weight at 25°C.

Detailed Description of the Invention

30

The active herbicidal compound or compounds within the non aqueous or "oil" phase of the present invention are preferably non-water miscible herbicides which are liquid below 30°C or non-water miscible 35 herbicides which are a solid above 30°C and are soluble

in non aqueous solvents commonly employed as herbicidal carriers at levels above 30 percent by weight at 20°C. Examples of non-water miscible herbicides which are a liquid below 30°C are:

5

3,5,6-trichloro-2-pyridinyloxyacetic acid as the butoxyethyl ester triclopyr butoxyethyl ester;(4-chloro-2-methyl phenoxy) acetic acid as the butoxyethyl or iso-octyl esters (MCPA-butoxy ethyl ester and MCPA-iso-octyl ester);

10

(±)-2-4-(4-chloro-2-methylphenoxy)propionic acid as the iso-octyl ester (Mecoprop-iso-octyl ester);

15

3,6-dichloro pyridine-2-carboxylic acid as the iso-octyl ester (clopyralid iso-octyl ester);

20

(±)-2-[4-[[5-(trifluoromethyl)-2-pyridil]-oxy]phenoxy] propionic acid as the butyl ester (fluazifop-butyl);

25

(R)-2-[4-(5-trifluoromethyl)-2-pyridinyl-oxy]phenoxy] propionic acid as the butyl ester (fluazifop-P-butyl);

30

(2,4-dichlorophenoxy)acetic acid, (2,4-D) as the 2-butoxyethyl, iso-octyl or butyl esters, (2,4-D-butoxyethyl ester, 2,4-D-iso-octyl ester and 2,4-D-butyl ester);

35

(±)-2-(2,4-dichlorophenoxy)propionic acid, 2,4-DP as the iso-octyl, 2-butoxyethyl esters (2,4-DP-iso-octyl ester and 2,4-DP 2-butoxyethyl ester); and

O-(6-chloro-3-phenyl-4-pyridazinyl)-S-octyl
carbanothioate (pyridate);

5 all of which are commercially available. Preferably,
the liquid non-water miscible herbicide is trichlopyr
oran ester thereof such as triclopyr butoxyethyl ester
(available from DowElanco under the Trade Mark
CROSSBOW).

10 Examples of non-water miscible herbicides
which are a solid above 30°C are:

[4-amino-3,5-dichloro-6-fluoro-2-pyridinyl]]
acetic acid, as 1-methyl heptyl ester (fluoxypry-
15 1-methyl heptyl ester);

N-benzoyl-N-(3,4-dichlorophenyl)-DL-alanine ethyl
ester (benzoyl pro-ethyl);

20 2,6-dichloro-4-cyanophenyl octanoate (bromoxynil
octanoate);

2-[1-(ethoxyimino)butyl]-3-hydroxy-5-(tetrahydro-
2H-thiopyran-3-yl)-2-cyclohexane-1-one
25 (cycloxydim);

S-(2,3-dichloro-2-propenyl)bis(1-methylethyl)
carbamothioate (di-allate);

30 (RS-2-[4-(2,4- dichlorophenoxy)phenoxy]propanoic
acid, methyl ester (diclofop methyl);

(±)-2-[4-[(6-chloro-2- benzoxazolyl)oxy]phenoxy]
propanoic acid, ethyl ester (fenoxaprop ethyl);
35

N-benzoyl-N-(3-chloro-4-fluorophenyl)-D-alanine,
methyl and isopropyl esters (flamprop-M-isopropyl
and flamprop-M-methyl);

5 (±)-2-[4-[(3-chloro-5-(trifluoromethyl)-2-
pyridinyl]oxy]phenoxy]propionic acid, as methyl
and ethoxy ethyl esters (haloxyfop- methyl and
haloxyfop-ethoxyethyl);

10 2,6-diiodo-4-cyanophenyl octanoate (ioxynil
octanoate);

N-(1-ethylpropyl)-3,4-dimethyl-2,6-dinitro
benzenamine (pendimethalin) and

15 (±)-2-[4-[(6-chloro-2-quinoxalinyloxy)-
phenoxy]propanoic acid as ethyl ester
(Quizalofop-ethyl);

20 all of which are commercially available.

Preferably, the non-water miscible herbicide
which is a solid above 30°C is fluroxypyr, or an ester
thereof. Fluroxypyr is described in U.S. Patent

25 3,761,486 and is represented by the formula

30

Esters of fluroxypyr are disclosed in U.S.
Patent 3,755,339. Fluroxypyr-1-methyl heptyl ester is
35 available from DowElanco under the Trade Mark STARANE.

The water-insoluble herbicides, such as fluroxypyr or triclopyr will be present in the emulsion composition at a concentration of from about 1 to about 30 percent by weight, preferably from about 2 to about 20 percent by weight, and more preferably from about 2 to about 15 percent by weight.

Any organic solvent in which the fluroxypyr or triclopyr are soluble, and which is non-reactive with glyphosate and which is essentially water-insoluble (solubility less than 1 percent by weight) may be employed in the emulsions of the present invention. Suitable organic solvents include, for example, xylene, kerosene, C₉ aromatics, N-alkylpyrrolidines such as N-octyl or N-dodecyl esters derived from naturally occurring fatty acids, monochlorobenzene and the like, or a blend of two or more such materials. Organic solvents which have a high flash point and which are of low toxicity to man will be preferably utilized in the emulsion compositions of the present invention. The organic solvent is present in the emulsion composition at a concentration of from 2 to 25 percent by weight, preferably at from 5 to 20 percent by weight of the total emulsion composition.

The water-soluble herbicide active ingredient of the present invention is a salt of N-phosphonomethyl glycine (glyphosate). Suitable salts of glyphosate are those selected from the group consisting of Group I and Group II metals having an atomic number up to 30, hydrochloride, pyridine, ammonium, aniline, lower aliphatic hydrocarbon amines and lower alkanol amines, wherein "lower" designates aliphatic radicals which have up to 4 carbon atoms in

a straight or branched chain. The preferable salt of glyphosate for use in the present invention is the isopropylamine salt. The concentration of the glyphosate salt present in the emulsion of the present invention is from 1 to 30 percent by weight, preferably from 10 to 25 percent by weight of the total composition. The isopropylamine salt of N-phosphonomethyl glycine is commercially available from Monsanto Company as a concentrate under the Trade Mark ROUNDUP.

The emulsifying system essential to the present invention is a blend of amine-ethoxylate surfactants which create the proper hydrophile to lipophile balance (HLB) to maintain the herbicides of the present invention as an emulsion. According to the HLB system, surfactants are classified by the size and strength of the hydrophilic and lipophilic portions of the molecule. The balance in the size and strength of these two opposing groups is called the hydrophile-lipophile balance or HLB. An emulsifier that is lipophilic in character is assigned a low HLB number, while an emulsifier that is hydrophilic in character is assigned a high number. The values that have been assigned to emulsifiers range from one to forty with the midpoint being about ten. A thorough explanation of hydrophilic-lipophilic balance can be found in L. I. Osipov, Surface Chemistry (1962).

Suitable ethoxylated amine surfactants for use in the present invention include, for example, ethoxylated primary amines derived from animal and vegetable fatty acids; ethoxylated synthetic C₁₂-C₁₈ primary, secondary or tertiary amines; ethoxylated di- and polyamines; polyoxyethylenated alkylamines, quaternized polyoxyethylenated amines (eg. alkylamines) and alkyldimethylamine oxides.

Particularly useful are ethoxylated amines such as oleylamine, cocamine or tallow amine, all with 1 to 20 moles of ethylene oxide; and hydrogenated tallow amines with 1 to 20 moles ethylene oxide. The total
5 amount of emulsifier will generally be in the range of from 1 to 30 percent, preferably from 2 to 25 percent, and more preferably from 2 to 15 percent by weight of the total composition. The preferred combination of
10 surfactants being an amineethoxylate containing 2 to 5 moles of ethylene oxide and being a cocoamine ethoxylate containing between 10 to 20 moles of ethylene oxide.

Commercially available ethoxylated amines
15 are generally a mixture of ethoxy chain lengths. For example, a commercial product which is nominally 2 mole ethoxylated will normally contain small amounts of a product which is 1 or 3 mole ethoxylated. As the degree of ethoxylation increases, the "spread" of
20 ethoxylate chain length also increases. However, the spread of ethoxylate chain length in such commercial products is not generally very broad, and a material with a nominal ethoxylate content of 15 moles of ethylene oxide per molecule will not normally contain
25 any significant amount of material with less than 13 or more than 17 ethylene oxide units.

The ethoxylated amine surfactants employed
in the present invention are commercially available
30 from a number of manufactures as described in McCutcheon's Emulsifiers and Detergents", North American Edition, 1989, McCutcheon Division, MC Publishing Co., 175 Rock Rd., Glen Rock, N.J. 07452, U.S.A.

35

The emulsifier blend is such as to produce a microemulsion wherein the droplet size is between 5 and 500 nanometers, preferably between 5 and 250 nanometers, and more preferably between 5 and 100 nanometers.

The present invention also requires the presence of a cosurfactant to provide the proper balance between the hydrophilic and lipophilic properties for the required oil and water phases under the conditions of use. Cosurfactants suitable for use in the present invention are, for example, primary C₄-C₁₂ aliphatic alcohols, such as butanol and nonanol; alcohol ethers based on ethylene, propylene or butylene oxides such as, 2-butoxy ethanol and dipropylene glycol monomethylether; and nonionic surfactants with HLB values less than 10. Examples of suitable nonionic surfactants are, for example, C₉-C₁₆ alcohol ethoxylates and block copolymers of propylene oxide or ethylene oxide. Other suitable nonionic surfactants include alkyl phenol ethoxylates having 1 to 5 moles of ethylene oxide such as nonyl phenol with 1 mole of ethylene oxide. The amount of cosurfactant in the present invention is from 0.5 to 10 percent by weight, preferably 2 to 5 percent by weight of the total composition.

Suitable cosurfactants are commercially available, such as dipropylene glycol methyl ether, sold by The Dow Chemical Company under the Trade Mark DOWANOL.

In the emulsions of the present invention, water is present in varying amounts, depending upon the quantities employed of the other ingredients and

is present in sufficient quantity to serve as the continuous phase for the water-insoluble dispersed herbicide component.

5 The herbicidal emulsions of the present invention can contain minor quantities of one or more inert formulation adjuvants, such as antifoaming agents, anti-caking agents, biocides, dyes, anti-corrosion agents, freeze point depressants and the
10 like, especially if the compositions are to be stored for any extended period of time prior to use, particularly under adverse storage conditions.

 Preparation of the emulsion composition
15 described herein can be carried out using standard procedures in the industry. Generally, the water-insoluble herbicide is first dissolved in the organic solvent and then blended with the emulsifying surfactants. Glyphosate isopropylamine concentrate
20 (Trade Mark ROUNDUP) is then blended under low shear with the solution containing the water-insoluble herbicide. The cosurfactant is then added followed by water to bring the composition to the desired concentration. The complete mixture is then blended
25 until homogeneity is achieved. Advantageously, when using an herbicide such as fluroxypyr, which is a solid below 30°C, the herbicide is added in the molten state to the organic solvent.

30 An embodiment of the invention is illustrated by the following Example.

Example I

The following composition is an example of the formulation of the present invention.

| <u>Ingredient</u> | <u>Grams/Liter</u> | <u>Percent</u> <u>w/w</u> |
|--|--------------------|------------------------------|
| Fluroxypyr-1-methylheptyl ester | 72 | 6.58 |
| C ₉ aromatic solvent (Trade Mark SOLVESSO 150) | 118 | 10.05 |
| Glyphosate - isopropylamine salt | 250 | 22.86 |
| 2 mole ethoxylated oleyl amine (Trade Mark ETHOMEEN S-12) | 62 | 5.67 |
| 15 mole ethoxylated cocoamine (Trade Mark ETHOMEEN C-25) | 38 | 3.47 |
| Dipropylene glycol monomethylether (Trade Mark DOWANOL DPM) | 30 | 2.74 |
| Water | Balance to 1 liter | Balance |

The above formulation was prepared in the following manner:

A. Molten fluroxypyr-1-methylheptyl ester was dissolved in Solvesso 150, and then blended with the two surfactants, Ethomeen S-12 and C-25.

B. The solution prepared in (A) was added to a glyphosate isopropylamine concentrate. The glyphosate isopropylamine concentrate was added as 614g of the commercial herbicidal concentrate ROUNDUP.

C. Dowanol DPM was then added to the mixture in B above followed by water and the complete mixture blended until homogeneity was achieved.

5 Example IA

10 The chemical stability of an emulsion formulation as prepared in Example I under various storage temperatures is given in Table IA. The emulsion samples were stored in glass and the data given as acid equivalents of the fluroxypyr acid as measured by high performance chromatography using a 24 cm x 4.6 mm column coated with APEX ODS (5 micron), stationary phase eluting with 65:35 acetonitrile:water. The data
15 indicates only up to about a 3 percent loss after one month storage at 55°C.

TABLE IA
Chemical Stability

| Storage period | Storage temperature | Grams Fluroxypyr (Starane) Acid Equivalents/Liter |
|----------------|---------------------|---|
| One month | -10°C | 47.1 |
| | 40°C | 46.6 |
| | 55°C | 45.6 |

Example IB

The dilution characteristics of the emulsion formulation as prepared in Example I in soft (20 ppm CaCO_3) and hard (500 ppm CaCO_3) waters as defined by the

World Health Organization are given in Table IB, and after storage for one month at various temperatures in Table IC. The dilution test was done using a 2 percent v/v dilution in 20 and 500 ppm CaCO_3 equivalents water at 20°C utilizing the Collaborative International Pesticides Analytical Committee (CIPAC) test method, MT36.1.

TABLE IB
Dilution Characteristics

| Time | 20 ppm CaCO_3 | 500 ppm CaCO_3 |
|---------------|------------------------|-------------------------|
| 1/2 hour | Trace ¹ | Nil ² |
| 1 hour | Trace | Nil |
| 2 hours | Trace | Nil |
| Re-dispersion | Complete | Complete |
| 2½ hours | Nil | Nil |

¹Trace means less than 0.05 percent by volume cream.

²Nil means no visible cream.

TABLE IC

Dilution Characteristics after Storage for one month

| Time | Storage at -10°C | | Storage at 40°C | | Storage at 55°C | |
|---------------|------------------|----------|-----------------|----------|-----------------|----------|
| | 20 | 500 | 20 | 500 | 20 | 500 |
| ½ hour | Nil* | Nil | Nil | Nil | Nil | Nil |
| 1 hour | Nil | Nil | Nil | Nil | Nil | Nil |
| 2 hours | Nil | Nil | Nil | Nil | Nil | Nil |
| Re-dispersion | Complete | Complete | Complete | Complete | Complete | Complete |
| 2½ hours | Nil | Nil | Nil | Nil | Nil | Nil |

Nil* means no visible cream

Example ID

The variation in the pH of an emulsion formulation as prepared in Example I after storage in glass under various conditions is given in Table 1D.

TABLE ID
Variation of pH

| Store Time | Store Temperature | Neat | 5 Percent (v/v) Dilution |
|------------|-------------------|------|--------------------------|
| Initial | - | 5.76 | 5.08 |
| 1 month | -10°C | 6.35 | 5.48 |
| | 40°C | 6.29 | 5.63 |
| | 55°C | 6.26 | 5.58 |

Example II

Microemulsion containing Haloxyfop ethoxyethyl ester and Glyphosate as the isopropylamine salt.

A microemulsion preparation was prepared utilising the following ingredients:-

| Composition | | |
|--|---------|-------|
| Ingredient | g/Litre | % w/w |
| Haloxyfop ethoxy ethyl ester | 35.0 | 3.19 |
| C9 Aromatic solvent (Trade Mark Solvesso 150) | 65.0 | 5.92 |

| <u>Composition</u> | | |
|--|-----------------------|--------------|
| <u>Ingredient</u> | <u>g/Litre</u> | <u>% w/w</u> |
| Glyphosate (as isopropylamine salt) | 225 | 20.48 |
| 5 mole ethoxylated cocoamine (Trade Mark Ethomeen C-15) | 50.0 | 4.55 |
| 15 mole ethoxylated cocoamine (Trade Mark Ethomeen C-25) | 250 | 22.76 |
| Dipropylene glycol monomethyl ether (Trade Mark Dowanol DPM) | 60 | 5.46 |
| Water | Balance to 1 litre | Balance |

The formulation was prepared in the following manner. Molten Haloxyfop ethoxyethyl ester was dissolved in Solvesso 150 and then blended with the two surfactants, Ethomeens C-15 and C-25. The resulting solution was added to a glyphosate isopropylamine concentrate. The glyphosate concentrate was added as 553g of the commercial herbicidal concentrate, ROUNDUP. DOWANOL DPM was then added to the mixture above, followed by water and the complete mixture blended until homogeneity was achieved.

Example III

Microemulsion concentrate containing triclopyr butoxy-ethyl ester and Glyphosate as the isopropylamine salt.

A microemulsion composition was employed utilising the following ingredients:-

| <u>Composition</u> | | |
|---|-----------------------|--------------|
| <u>Ingredient</u> | <u>g/Litre</u> | <u>% w/w</u> |
| Triclopyr butoxy ethyl ester | 139 | 12.32 |
| Glyphosate (as Isopropylamine salt) (Trade Mark Solvesso 150) | 225 | 19.94 |
| 5 mole ethoxylated cocoamine (Trade Mark Ethomeen C-15) | 50.0 | 4.43 |
| 15 mole ethoxylated cocoamine (Trade Mark Ethomeen C-25) | 235 | 20.82 |
| Dipropylene glycol monomethyl ether (Trade Mark Dowanol DPM) | 50 | 4.43 |
| Water | Balance to 1 litre | Balance |

The formulation was prepared in the following manner. Ethomeens C-15 and C-25 were blended with triclopyr butoxy ester. The resulting solution was added to 553 grams of the commercial herbicidal concentrate, ROUND-UP. DOWANOL DPM was then added to the mixture followed by water and the complete mixture blended until homogeneity was achieved.

Example IV

Microemulsion concentrate containing 2,4-D

iso-octyl ester and glyphosate as the isopropylamine salt.

A microemulsion was prepared, utilising the following ingredients:-

| <u>Composition</u> | | |
|--|-----------------------|--------------|
| <u>Ingredient</u> | <u>g/Litre</u> | <u>% w/w</u> |
| 2,4-D iso octyl ester | 151 | 13.57 |
| Glyphosate (as Isopropylamine salt) | 225 | 20.22 |
| 2 mole ethoxylated cocoamine (Trade Mark Ethomeen C-12) | 40 | 3.59 |
| 15 mole ethoxylated cocoamine (Trade Mark Ethomeen C-25) | 320 | 28.76 |
| Dipropylene glycol monomethyl ether (Trade Mark Dowanol DPM) | 60 | 5.39 |
| Water | Balance to 1 litre | Balance |

The formulation was prepared in the following manner. Ethomeens C-12 and C-25 were blended with 2,4-D iso-octyl ester. The resulting solution was added to 553 grams of the commercial herbicidal concentrate, ROUND-UP. DOWANOL DPM was then added to the mixture followed by water and the complete mixture blended until homogeneity was achieved.

The sprayable herbicide composition formulated from the microemulsion concentrate from this invention may be applied from all types of

presently used equipment with no plugging of the spray nozzle or other malfunction of the spray equipment.

The herbicide emulsions described herein produce weed control comparable to the conventional tank mixtures of the active herbicidal ingredients. When the emulsion of Example 1 was compared with a comparable tank mix containing active ingredients sourced from commercial products, no significant differences in weed control were observed.

As can be seen from the foregoing, the preferred embodiments of this invention provide a flowable, herbicidal microemulsion concentrate composition containing the water-soluble isopropylamine salt of N-phosphonomethylglycine (glyphosate) and a water-insoluble fluroxypyr or triclopyr herbicide within a novel emulsifier system as a pre-packaged, one- container mixture which can be conveniently stored and handled and which has enhanced storage-stability.

Other embodiments of the invention will be apparent to those skilled in the art from a consideration of this specification or practice of the invention disclosed herein. It is intended that the Examples be considered as exemplary only, with the scope of the invention being indicated by the following claims.

CLAIMS

1. A water-dispersible herbicidal microemulsion concentrate composition comprising:

A. from 1 to 30 percent by weight of at least one water-insoluble herbicide;

B. optionally, from 0 to 25 percent by weight of a non-water miscible organic solvent in which the water-insoluble herbicide is soluble;

C. from 1 to 30 percent by weight of a water-soluble salt of glyphosate;

D. from 0.5 to 35 percent by weight of at least one amine ethoxylate surfactant containing from 2 to 5 moles of ethylene oxide;

E. from 0.5 to 35 percent by weight of at least one amine ethoxylate surfactant containing from 8 to 20 moles of ethylene oxide;

F. from 0.5 to 10 percent by weight of at least one cosurfactant which is a glycol ether, a C₄-C₁₂ aliphatic alcohol, an alcohol ether based on ethylene, propylene or butylene oxide, or a nonionic surfactant with an HLB value of less than 10; and,

G. sufficient water to serve as the continuous phase for the emulsion.

2. A composition as claimed in Claim 1, wherein the said at least one water-insoluble herbicide is at least one of fluroxypyr, triclopyr or an ester thereof.

3. A composition as claimed in Claim 2, wherein the water-insoluble herbicide is fluroxypyr-1-methylheptyl ester.

4. A composition as claimed in any one of the preceding Claims, wherein the salt of glyphosate is the isopropylamine salt.

5. A composition as claimed in any one of the preceding claims, wherein the amount of component D is from 0.5 to 10 percent by weight of the composition.

6. A composition as claimed in any one of the preceding claims, wherein the amount of component E is from 0.5 to 10 percent by weight of the composition.

7. A composition as claimed in Claim 1, comprising:

A. from 2 to 20 percent by weight of fluroxypyr 1-methylheptyl ester;

B. from 5 to 20 percent by weight of a non-water miscible organic solvent in which the fluroxypyr is soluble;

C. from 10 to 25 percent by weight of the isopropylamine salt of glyphosate;

D. from 0.5 to 10 percent by weight of at least one amine ethoxylate containing 2 moles of ethylene oxide;

E. from 0.5 to 10 percent by weight of at least one cocamine ethoxylate surfactant containing from 10 to 20 moles of ethylene oxide;

F. from 2 to 5 percent by weight of at least one cosurfactant which is a glycol ether, a C₄-C₁₂ aliphatic alcohol, an alcohol ether based on ethylene, propylene or butylene oxide, or a nonionic surfactant with an HLB value of less than 10; and,

G. sufficient water to serve as the continuous phase for the emulsion.

8. A composition as claimed in Claim 7, consisting essentially of

A. from 2 to 20 percent by weight of fluroxypyr 1-methylheptyl ester;

B. from 5 to 20 percent by weight of a non-water miscible organic solvent in which the fluroxypyr is soluble;

C. from 10 to 25 percent by weight of the isopropylamine salt of glyphosate;

D. from 0.5 to 10 percent by weight of at least one amine ethoxylate containing 2 moles of ethylene oxide;

E. from 0.5 to 10 percent by weight of at least one cocamine ethoxylate surfactant containing from 10 to 20 moles of ethylene oxide;

F. from 2 to 5 percent by weight of at least one cosurfactant which is a glycol ether, a C₄-C₁₂ aliphatic alcohol, an alcohol ether based on ethylene, propylene or butylene oxide, or a nonionic surfactant with an HLB value of less than 10.

9. A herbicidal composition prepared by diluting a herbicidal microemulsion concentrate composition as claimed in any one of the preceding claims to a dilution suitable for application to weeds.

10. A method for the control or prevention of weed infestation, which method comprises applying to the weeds, or to a locus in which the growth of weeds is to be prevented, a composition as claimed in Claim 9.

Patents Act 1977
Examiner's report to the Comptroller under
Section 17 (The Search Report)

-26-

Application number
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Relevant Technical fields

(i) UK CI (Edition K) A5E EF ES
(ii) Int CL (Edition 5) A01N

Search Examiner
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Databases (see over)

(i) UK Patent Office
ONLINE DATABASES: WPI
(ii)

Date of Search
24 JUNE 1992

1-10

Documents considered relevant following a search in respect of claims

| Category (see over) | Identity of document and relevant passages | Relevant to claim(s) |
|------------------------|--|-------------------------|
| | NONE | |

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SF2(b)

| Category | Identity of document and relevant passages | Relevant to claim(s) |
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Categories of documents

X: Document indicating lack of novelty or of inventive step.

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